

Form PTO-1390 (Rev. 12-29-99) <b>TRANSMITTAL LETTER TO THE UNITED STATES          DESIGNATED/ELECTED OFFICE (DO/EO/US)          CONCERNING A FILING UNDER 35 U.S.C. 371</b>		US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE ATTORNEY'S DOCKET NO. <b>H 3876 PCT/US</b> U.S. APPLICATION NO. (if known see 37 CFR 1.5) <b>09/913377</b>
INTERNATIONAL APPLICATION NO. <b>PCT/EP00/00903</b>	INTERNATIONAL FILING DATE <b>February 4, 2000</b>	PRIORITY DATE CLAIMED <b>February 13, 1999</b>
TITLE OF INVENTION <b>METHOD FOR PRODUCING PHYTOSTEROLS</b>		
APPLICANT(S) FOR DO/EO/US <b>Joerg Schwarzer, Bernhard Gutsche</b>		
Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:		
1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371. 3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1). 4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)). a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). b. <input type="checkbox"/> have been transmitted by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). (UNEXECUTED) 10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). Items 11 to 16. below concern other document(s) or information included: 11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input checked="" type="checkbox"/> A FIRST preliminary amendment <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 14. <input type="checkbox"/> A substitute specification. 15. <input type="checkbox"/> A change of power of attorney and/or address letter. 16. <input type="checkbox"/> Other items or information:		
<b>"Express Mail Post Office to Addressee" service Mailing Label Number  <u>EL541612567US</u></b>		

Form PTO 1390 (REV 12-29-99) page 2 of 2

"Express Mail " Mailing Label Number EL541612567US .

PATENT  
Docket No. H 3876 PCT/US

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

RE: PCT/EP00/00903  
International Filing Date: February 4, 2000  
Priority Date Claimed: February 13, 1999  
Applicant: Schwarzer, et al.  
Title: METHOD FOR PRODUCING PHYTOSTEROLS  
Applicants' Reference: H 3876 PCT/US

**PRELIMINARY AMENDMENT**

Commissioner for Patents  
Box PCT  
Washington, DC 20231

ATTN: DO/EO/US

Prior to the calculation of fees and examination of the above-identified national stage application pursuant to the accompanying submission under 35 U.S.C. §371, please amend the English translation of the International Application submitted herewith, without prejudice, as follows:

**In the Specification:**

Please amend the instant Specification, without prejudice, as follows:

Please delete all text above line 7 of page 1, including the heading "Prior Art", and replace the deleted matter with the following new section headings and title of the invention:

**--TITLE OF THE INVENTION**

Processes for Preparing Phytosterols and Substantially  
Citrostadienol-Free Phytosterols Prepared Thereby

**BACKGROUND OF THE INVENTION--**

At page 2, line 13 thereof, please delete the section heading "Description of the

**Preliminary Amendment of U.S. National Stage for International Application  
PCT/EP00/00903 filed February 4, 2000**

Invention" and insert the following new section heading and new paragraph:

**--BRIEF SUMMARY OF THE INVENTION**

The present invention relates, in general, to food additives and phytosterols for use therein, and more particularly, to a new process for the production of phytosterols substantially free from citrostadienol.--

At page 3, line 1 thereof, please insert the following new section heading:

**--DETAILED DESCRIPTION OF THE INVENTION--**

At page 6, between lines 1 and 2, please add the following new paragraph:

--What is claimed is:--.

On a separate, new page 7, following page 6, please add the following new section heading and paragraph containing an Abstract of the Disclosure:

**--ABSTRACT OF THE DISCLOSURE**

Processes for preparing substantially citrostadienol-free phytosterols are disclosed. The processes described include dissolving a liquid phytosterol in a hydrocarbon solvent, optionally with added methanol, and crystallizing the phytosterol compounds. Also described are the resulting phytosterols which are substantially citrostadienol-free, and preferably contain less than 0.5% by weight citrostadienol.--

**In the Claims:**

Please add new claims 9-37, as follows:

--9. (New) A process for preparing phytosterols, said process comprising:

- (a) providing a liquid phytosterol starting material obtained by transesterification of a distillation residue with an alkanol;
- (b) dissolving the liquid phytosterol starting material in a hydrocarbon

**Preliminary Amendment of U.S. National Stage for International Application  
PCT/EP00/00903 filed February 4, 2000**

solvent; and

(c) crystallizing a phytosterol product, wherein the phytosterol product is substantially citrostadienol-free.--

--10. (New) The process according to claim 9, wherein the distillation residue comprises a deodorizer condensate obtained from fatty acid methyl ester production.--

--11. (New) The process according to claim 10, wherein the deodorizer condensate is derived from an oil selected from the group consisting of rapeseed oil and sunflower oil.--

--12. (New) The process according to claim 11, wherein the oil comprises sunflower oil.--

--13. (New) The process according to claim 9, wherein the distillation residue comprises tall oil pitch.--

--14. (New) The process according to claim 9, wherein the alkanol comprises methanol.--

--15. (New) The process according to claim 11, wherein the alkanol comprises methanol.--

--16. (New) The process according to claim 9, wherein the liquid phytosterol starting material is maintained at a temperature of from 60°C to 80°C prior to and during dissolution in the hydrocarbon solvent.--

--17. (New) The process according to claim 11, wherein the liquid phytosterol starting material is maintained at a temperature of from 60°C to 80°C prior to and during

**Preliminary Amendment of U.S. National Stage for International Application  
PCT/EP00/00903 filed February 4, 2000**

dissolution in the hydrocarbon solvent.--

--18. (New) The process according to claim 16, wherein the liquid phytosterol starting material is maintained at a temperature of from 65°C to 70°C.--

--19. (New) The process according to claim 17, wherein the liquid phytosterol starting material is maintained at a temperature of from 65°C to 70°C.--

--20. (New) The process according to claim 9, wherein the hydrocarbon solvent comprises a linear or branched alkane isomer selected from the group consisting of pentane, hexane, heptane, octane, nonane, decane, and mixtures thereof.--

--21. (New) The process according to claim 9, wherein the hydrocarbon solvent comprises a linear or branched alkane isomer selected from the group consisting of hexane, heptane, and mixtures thereof.--

--22. (New) The process according to claim 9, wherein methanol is combined with the hydrocarbon solvent prior to crystallization.--

--23. (New) The process according to claim 22, wherein the methanol is present in an amount of from 1 to 15 % by weight, based on the hydrocarbon solvent.--

--24. (New) The process according to claim 11, wherein methanol is combined with the hydrocarbon solvent prior to crystallization.--

--25. (New) The process according to claim 24, wherein the methanol is present in an amount of from 1 to 15 % by weight, based on the hydrocarbon solvent.--

--26. (New) The process according to claim 16, wherein methanol is

**Preliminary Amendment of U.S. National Stage for International Application  
PCT/EP00/00903 filed February 4, 2000**

combined with the hydrocarbon solvent prior to crystallization.--

--27. (New) The process according to claim 26, wherein the methanol is present in an amount of from 1 to 15 % by weight, based on the hydrocarbon solvent.--

--28. (New) The process according to claim 9, wherein crystallizing the phytosterol product comprises cooling the liquid phytosterol starting material in the hydrocarbon solvent to a temperature of below about 30°C.--

--29. (New) The process according to claim 9, wherein crystallizing the phytosterol product comprises cooling the liquid phytosterol starting material in the hydrocarbon solvent to a temperature of from about 25°C to about 30°C.--

--30. (New) The process according to claim 11, wherein crystallizing the phytosterol product comprises cooling the liquid phytosterol starting material in the hydrocarbon solvent to a temperature of below about 30°C.--

--31. (New) The process according to claim 11, wherein crystallizing the phytosterol product comprises cooling the liquid phytosterol starting material in the hydrocarbon solvent to a temperature of from about 25°C to about 30°C.--

--32. (New) The process according to claim 9, wherein the phytosterol product has a citrostadienol content of less than 0.5% by weight.--

--33. (New) The process according to claim 9, wherein the phytosterol product has a citrostadienol content of less than 0.2% by weight.--

--34. (New) A process for preparing phytosterols, said process comprising:  
(a) providing a liquid phytosterol starting material obtained by

**Preliminary Amendment of U.S. National Stage for International Application  
PCT/EP00/00903 filed February 4, 2000**

transesterification of a distillation residue with methanol, wherein the distillation residue comprises a deodorizer condensate derived from sunflower oil;

(b) dissolving the liquid phytosterol starting material in a hydrocarbon solvent, the hydrocarbon solvent comprising a linear or branched alkane isomer selected from the group consisting of hexane, heptane, and mixtures thereof, wherein the liquid phytosterol starting material is maintained at a temperature of from 60°C to 80°C prior to and during dissolution in the hydrocarbon solvent; and

(c) crystallizing a phytosterol product via cooling the liquid phytosterol starting material in the hydrocarbon solvent to a temperature of below about 30°C, wherein methanol is combined with the hydrocarbon solvent prior to crystallization in an amount of from 1 to 15 % by weight, based on the hydrocarbon solvent, and wherein the phytosterol product has a citrostadienol content of less than 0.5% by weight.--

--35. (New) A phytosterol prepared by the process according to claim 1.--

--36. (New) A phytosterol prepared by the process according to claim 34.--

--37. (New) A composition comprising one or more natural phytosterol compounds, wherein the composition has a citrostadienol content of 0.5% by weight or less.--

Please cancel claims 1-8, without prejudice.



**Preliminary Amendment of U.S. National Stage for International Application  
PCT/EP00/00903 filed February 4, 2000**

**REMARKS**

Claims 9-37 are currently pending in the instant application.

The Specification has been amended to delete the original section headings and to insert the preferred section headings pursuant to 37 C.F.R. §1.77. A new Title of the Invention has been inserted. An Abstract of the Disclosure, in accordance with the disclosure, has been added. It is submitted that the amendments to the Specification made herein introduce no new matter. All of the amendments to the Specification constitute deletions of original section headings and/or paragraphs, and insertions or additions of new section headings and/or paragraphs. Accordingly, pursuant to 37 C.F.R. §1.121(b)(1)(iii), no separate page captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE" is necessary. A separate page containing a clean copy of the Abstract of the Disclosure has been attached for the Examiner's convenience. Entry of the amendments to the Specification made herein are therefore proper and respectfully requested.

Original claims 1-8 have been canceled and replaced with new claims 9-37 solely for the purpose of improving clarity and grammar, which may suffer in translation, and not for any reason which relates to the statutory requirements for a patent. New claims 9-37 have not been added in response to any rejection, nor in anticipation of any rejection. Applicants respectfully submit that the scope of new claims 9-37 generally corresponds to the scope of original claims 1-8, and that new claims 9-37 are no narrower than original claims 1-8. Furthermore, although a moot point in view of their cancellation, Applicants respectfully submit that original claims 1-8 satisfied the requirements of 35 U.S.C. §112, as filed. New claims 9-37 are supported by the claims as originally filed and in the Specification, for example, at page 2, lines 14-30; at page 3, lines 5-9 & 14-30; and in the Examples. No new matter has been introduced. All of the amendments to the Claims constitute cancellation of original claims and the addition of new claims. Accordingly, pursuant to 37 C.F.R. §1.121(c)(1)(ii), no separate page captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE" is necessary. Entry is therefore proper and respectfully requested.

**Preliminary Amendment of U.S. National Stage for International Application  
PCT/EP00/00903 filed February 4, 2000**

Prompt examination of the instant application in view of the amendments made  
herein is respectfully requested.

Respectfully submitted,

**JOERG SCHWARZER, et al.**

August 13, 2001  
(Date)

Aaron R. Ettelman  
AARON R. ETTELMAN  
(Reg. No. 42,516)  
Attorney for Applicants  
Telephone: (610) 278-4930  
Facsimile: (610) 278-6548  
E-Mail: AARON.ETTELMAN@HENKEL-AMERICAS.COM

Cognis Corporation, Patent Dept.  
2500 Renaissance Blvd., Suite 200  
Gulph Mills, PA 19406

ARE/ras

G:\Data\amend\H3876.pam.doc

## Method for Producing Phytosterols

### Field of the Invention

This invention relates generally to food additives and more particularly to a new process for the production of phytosterols substantially free from citrostadienol.

5

### Prior Art

Phytosterols and their esters possess hypocholesterolaemic properties, i.e. these substances are capable of lowering the cholesterol level in the blood. Accordingly, they are used as food additives, for example for the production of margarine, frying oils, sausage, ice cream and the like. The production of sterols and other unsaponifiable constituents, such as tocopherols for example, from distillates obtained in the deacidification of vegetable oils, has already been variously described in the literature, cf. **EP-A2 0 610 742** (Hoffmann-LaRoche), **GB-A1 2,145,079** (Nisshin Oil Mills Japan) and **EP-A1 0 333 472** (Palm Oil Research and Development Board).

European Patent **EP-B1 0 656 894** (Henkel) describes a process for the production of sterols in which a residue from the distillation of methyl esters consisting essentially of glycerides, sterols, sterol esters and tocopherols is transesterified with methanol in the presence of alkaline catalysts. After neutralization of the catalyst, removal of the excess methanol by distillation and, optionally, removal of the catalyst by washing, the sterols are crystallized by lowering the reaction temperature from about 65 to 20°C. The crystals obtained are then washed with methanol and water. However, where residues from the production of methyl esters based on sunflower oil are used, the sterols obtained contain not only the

target components, such as above all campesterol, campestanol, stigmasterol,  $\beta$ -sitosterol and  $\beta$ -sitostanol, but also significant amounts of citrostadienol which is undesirable for applicational reasons. German patent application **DE-A1 3226225** (Raisio) describes a process by which  
5 the amount of citrostadienol can be reduced. In this process, the solid sterols are first dissolved in heptane and then re-crystallized after addition of methanol. However, the resulting products are by no means free from citrostadienol and, in addition, the yields are unsatisfactory.

Accordingly, the problem addressed by the present invention was to  
10 provide high yields of phytosterols which would be distinguished above all by the fact that they would be largely free from citrostadienol.

### **Description of the Invention**

The present invention relates to a process for the production of  
15 phytosterols by alkali-catalyzed transesterification of residues from the production of methyl esters with methanol, neutralization of the catalyst and removal of the unreacted alcohol, characterized in that the transesterification products are dissolved in saturated hydrocarbons containing 5 to 10 carbon atoms at a temperature at which they are present  
20 in liquid form, the phytosterols are crystallized in the hydrocarbon by lowering the temperature, optionally after the addition of an adequate quantity of aqueous methanol, and are then removed and purified in known manner by filtration, washing and drying.

It has surprisingly been found that crystallization of the sterols in  
25 hydrocarbons coupled with the addition of effective quantities of aqueous methanol gives products which have citrostadienol contents below 0.5% by weight and preferably below 0.2% by weight and which are therefore substantially free from this unwanted component. Another advantage is that, in contrast to known processes, the yields of sterol in the  
30 crystallization step are significantly higher.

### Transesterification

The production of a sterol-rich fraction by transesterification of residues from the deacidification of vegetable oils and subsequent working up can be carried out as described in EP-B1 0 656 894. Suitable starting materials are the distillation residues obtained, for example, as so-called deodorizer condensates in the production of fatty acid methyl esters based on rapeseed oil or, more particularly, sunflower oil. Tall oil pitch, more particularly pitch obtained from birch bark, is also suitable. Where it relates to the production of the sterol fractions, reference is comprehensively made to the document cited above.

### Crystallization

A key feature of the new process is that the products obtainable from the transesterification are dissolved in the hydrocarbons at a temperature at which they are still liquid. This is preferably the case at 60 to 80°C and more particularly at 65 to 70°C. Suitable solvents are lower alkanes, for example pentane, hexane, heptane, octane, nonane and decane. Included herein are both the linear hydrocarbons and the branched structural isomers derived therefrom and mixtures thereof. However, the use of hexane, heptane or mixtures thereof has proved to be particularly advantageous. After the sterols have dissolved, the temperature is reduced to such a value that the pure sterols crystallize. It has proved to be of advantage in this regard to add an effective quantity of aqueous methanol to the mixture. 1 to 25% by weight aqueous methanol solutions are normally used for this purpose, the quantity in which they are added - based on the hydrocarbons - typically being in the range from about 1 to 15% by weight. Although the crystallization process begins at a temperature as low as about 30°C, it has proved to be of advantage to lower the temperature to about 15 to 25°C. The phytosterols obtained are

then removed and purified in known manner, i.e. filtered off, washed free from soaps and then dried to constant weight. The resulting products have a citrostadienol content of less than 0.5% by weight and preferably less than 0.2% by weight.

5

### Examples

**Example 1.** 200 g of a distillation residue from the production of sunflower oil fatty acid methyl ester containing inter alia 15% by weight of glycerides and 28% by weight of free or bound sterols were introduced together with 10 78 g of methanol into a 1-liter three-necked condenser equipped with a stirrer and distillation head. 3.8 g of 30% by weight sodium methylate solution were then added to the mixture, followed by stirring for 4 h at 70°C. The alkaline catalyst was then neutralized by addition of 4.2 g of citric acid 15 dissolved in 19 g of methanol, the unreacted methanol was distilled off in vacuo and the residue was washed soap-free with water at 65°C. A mixture of 400 g of hexane, 26 g of methanol and 8 g of water was added to the crude product and the whole was cooled to 20°C. Removal of the mother liquor through a filter and drying of the residue left 41 g of sterols 20 which were free from citrostadienol.

**Example 2.** The procedure was as described in Example 1 except that a mixture of 200 g of heptane, 13 g of methanol and 4 g of water was added to 100 g of the transesterification product and the whole was cooled for 4 25 hours to 20°C. Filtration and drying left 19.4 g of sterols with a citrostadienol content of less than 0.2% by weight.

**Comparison Example C1.** The procedure was as described in Example 1 except that methanol was added to the transesterification product in a ratio 30 by weight of 1:1. On cooling to 20°C, the crystals precipitated and were

filtered off, washed with aqueous methanol and then dried. However, the resulting sterols still contained 4.7% by weight of citrostadienol. 100 g of this product were dissolved in heptane at 70°C and, after the addition of 20 g of methanol, the whole was again cooled for 4 hours to a temperature of 5 20°C. Filtration and drying left only 75 g of sterols still with a citrostadienol content of 4.2% by weight.

10  
20  
30  
40  
50  
60  
70  
80  
90  
100  
110  
120  
130  
140  
150  
160  
170  
180  
190  
200  
210  
220  
230  
240  
250  
260  
270  
280  
290  
300  
310  
320  
330  
340  
350  
360  
370  
380  
390  
400  
410  
420  
430  
440  
450  
460  
470  
480  
490  
500  
510  
520  
530  
540  
550  
560  
570  
580  
590  
600  
610  
620  
630  
640  
650  
660  
670  
680  
690  
700  
710  
720  
730  
740  
750  
760  
770  
780  
790  
800  
810  
820  
830  
840  
850  
860  
870  
880  
890  
900  
910  
920  
930  
940  
950  
960  
970  
980  
990

**CLAIMS**

1. A process for the production of phytosterols by alkali-catalyzed transesterification of residues from the production of methyl esters with methanol, neutralization of the catalyst and removal of the unreacted  
5 alcohol, characterized in that the transesterification products are dissolved in saturated hydrocarbons containing 5 to 10 carbon atoms at a temperature at which they are present in liquid form, the phytosterols are crystallized in the hydrocarbon by lowering the temperature and are then removed and purified in known manner by filtration, washing and drying.
- 10 2. A process as claimed in claim 1, characterized in that residues from the production of sunflower oil fatty acid methyl esters or tall oil pitch are used.
3. A process as claimed in claims 1 and/or 2, characterized in that the transesterification products are dissolved at 60 to 80°C.
- 15 4. A process as claimed in at least one of claims 1 to 3, characterized in that hexane, heptane or mixtures thereof is/are used as the solvent.
5. A process as claimed in at least one of claims 1 to 4, characterized in that an effective quantity of aqueous methanol is added during crystallization.
- 20 6. A process as claimed in at least one of claims 1 to 5, characterized in that 1 to 25% by weight aqueous methanol solutions are used.
7. A process as claimed in at least one of claims 1 to 6, characterized in that the aqueous methanol solutions are used in quantities of 1 to 15% by weight, based on the hydrocarbons.
- 25 8. A process as claimed in at least one of claims 1 to 7, characterized in that phytosterols with a citrostadienol content of less than 0.5% by weight are produced.



Type a plus sign (+) inside this box → ☐

<b>DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION</b>  <input type="checkbox"/> Declaration Submitted with Initial Filing    OR <input checked="" type="checkbox"/> Declaration Submitted after Initial Filing	U.S. Department of Commerce Patent and Trademark Office	Attorney Docket Number	H 3876 PCT/US
		First Named Inventor	SCHWARZER, Joerg
	COMPLETE IF KNOWN		
	Application Number	09/913,377	
	Filing Date	11/13/2001	
	Group Art Unit		
		Examiner Name	

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**METHOD FOR PRODUCING PHYTOSTEROLS**

the specification of which (Title of the Invention)

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) 02/04/2000 as United States Application Number or PCT International

Application Number PCT/EP00/00903 and was amended on (MM/DD/YYYY)  (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES NO
199 06 551.9	DE	02/13/1999	<input type="checkbox"/>	<input type="checkbox"/> <input checked="" type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>

☐ Additional foreign application numbers are listed on a supplemental priority sheet attached hereto:

I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.
		<input type="checkbox"/>

Burden Hour Statement: This form is estimated to take .4 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.

"Express Mail Post Office  
to Addressee" service  
Mailing Label Number  
EL541613911US

## DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP00/00903	02/04/2000	

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

☐ Firm Name  Customer Number or label   
OR  
☒ List Attorney(s) and/or agent(s) name and registration number below:

Name	Registration Number	Name	Registration Number
John E. Drach Steven J. Trzaska	<u>32,891</u> <u>36,296</u>	Aaron R. Ettelman Henry E. Millson, Jr.	<u>42,516</u> <u>18,980</u>


☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☒ Customer Number or label **23657** OR ☒ Fill in correspondence address below

Name	Aaron R. Ettelman		
Address			
Address			
City	State	ZIP	
Country	Telephone	610-278-4930	Fax 610-278-6548

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this unsigned

Given Name	Joerg	Middle Initial		Family Name	Schwarzer	Suffix e.g. Jr.	
Inventor's Signature					Date	20-Aug-2001	
Residence: City	Hilden	State		Country	Germany	Citizenship	Germany
Post Office Address	Kunibertstrasse 13						
Post Office Address							
City	40723 Hilden	State		Zip		Country	Germany
Applicant Authority							

☒ Additional inventors are being named on supplemental sheet(s) attached hereto

## DECLARATION

ADDITIONAL INVENTOR(S)  
Supplemental Sheet

Name of Additional Joint Inventor, if any:										<input type="checkbox"/> A petition has been filed for this unsigned inventor											
Given Name		Bernhard				Middle Initial				Family Name		Gutsche				Suffix e.g. Jr.					
Inventor's Signature		<i>B Bernhard Gutsche</i>								Date		<i>Aug. 22. 2001</i>									
Residence: City		Hilden				State				Country		Germany				Citizenship		Germany			
Post Office Address		Kalstert 96																			
Post Office Address																					
City		40724 Hilden				State				Zip				Country		Germany				Applicant Authority	
Name of Additional Joint Inventor, if any:										<input type="checkbox"/> A petition has been filed for this unsigned inventor											
Given Name						Middle Initial				Family Name						Suffix e.g. Jr.					
Inventor's Signature										Date											
Residence: City						State				Country						Citizenship					
Post Office Address																					
Post Office Address																					
City						State				Zip				Country						Applicant Authority	
Name of Additional Joint Inventor, if any:										<input type="checkbox"/> A petition has been filed for this unsigned inventor											
Given Name						Middle Initial				Family Name						Suffix e.g. Jr.					
Inventor's Signature										Date											
Residence: City						State				Country						Citizenship					
Post Office Address																					
Post Office Address																					
City						State				Zip				Country						Applicant Authority	
Name of Additional Joint Inventor, if any:										<input type="checkbox"/> A petition has been filed for this unsigned inventor											
Given Name						Middle Initial				Family Name						Suffix e.g. Jr.					
Inventor's Signature										Date											
Residence: City						State				Country						Citizenship					
Post Office Address																					
Post Office Address																					
City						State				Zip				Country						Applicant Authority	
Name of Additional Joint Inventor, if any:										<input type="checkbox"/> A petition has been filed for this unsigned inventor											
Given Name						Middle Initial				Family Name						Suffix e.g. Jr.					
Inventor's Signature										Date											
Residence: City						State				Country						Citizenship					
Post Office Address																					
Post Office Address																					
City						State				Zip				Country						Applicant Authority	
<input type="checkbox"/> Additional inventors are being named on supplemental sheet(s) attached hereto																					

Please type a plus sign (+) inside this box 

PTO/SB/123 (10-00)  
Approved for use through 10/31/2002. OMB 0651-0035  
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

## CHANGE OF CORRESPONDENCE ADDRESS *Patent*

Address to:  
Assistant Commissioner for Patents  
Washington, D.C. 20231

Patent Number

Issue Date

Application Number

Filing Date

First Named Inventor

09/913,377

11/13/2001

Schwarzer, Joerg

Please change the Correspondence Address for the above-identified patent to:

☒ Customer Number

23657

Type Customer Number here

Place Customer  
Number Bar Code  
Label here

OR

☐

Firm or  
Individual Name

Cognis Corporation

Address

Address

City

State

ZIP

Country

Telephone

(610) 278-4920

Fax

(610) 278-6548

This form cannot be used to change the data associated with a Customer Number. To change the data associated with an existing Customer Number use "Request for Customer Number Data Change" (PTO/SB/124).

This form will not affect any "fee address" provided for the above-identified patent. To change a "fee address" use the "Fee Address Indication Form" (PTO/SB/47).

I am the :

☐

Patentee.

☐

Assignee of record of the entire interest. See 37 CFR 3.71.  
Statement under 37 CFR 3.73(b) is enclosed. (Form PTO/SB/96).

☒

Attorney or agent of record.

Typed or  
Printed Name

Aaron R. Ettelman, R.N. 42,516

Signature

Date

November 13, 2001

NOTE: Signatures of all the inventors or assignees of record of the entire interest or their representative(s) are required. Submit multiple forms if more than one signature is required, see below\*.

☐ \*Total of \_\_\_\_\_ forms are submitted.

Burden Hour Statement: This form is estimated to take 3 minutes to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.